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Published in:
Journal of Environmental Radioactivity

Link to article, DOI:
10.1016/j.jenvrad.2016.02.011

Publication date:
2016

Document Version
Peer reviewed version

Iodine isotopes in precipitation: four-year time series variations before and after 2011 Fukushima nuclear accident

Sheng Xu[1,3,*], Luyuan Zhang[2], Stewart P.H.T. Freeman[1], Xiaolin Hou[2], Akira Watanabe[3], David C.W. Sanderson[1], Alan Cresswell[1,3] and Katsuhiko Yamaguchi[3]

[1]Scottish Universities Environmental Research Center
East Kilbride, G75 0QF, UK

[2]Center for Nuclear Technologies, Technical University of Denmark
4000 Roskilde, Denmark

[3]Fukushima University, Fukushima 960-1296, Japan

[*]Corresponding author
Fax: +44 1355 270189; E-mail: s.xu@suerc.gla.ac.uk

Abstract

Rainwater samples were collected monthly from Fukushima, Japan, in 2012-2014 and analysed for ^{127}I and ^{129}I. These are combined with previously reported data to investigate atmospheric levels and behaviour of Fukushima-derived ^{129}I before and after the 2011 nuclear accident. In the new datasets, ^{127}I and ^{129}I concentrations between October 2012 and October 2014 varied from 0.5 to 10 μg/L and from 1.2×10^{8} to 6.9×10^{9} atom/L respectively, resulting in $^{129}I/^{127}I$ atomic ratio ranges from 3×10^{-8} to 2×10^{-7}. ^{127}I concentrations were in good agreement with those in the previous period from March 2011 to September 2012, whereas the ^{129}I concentrations and $^{129}I/^{127}I$ ratios followed declining trends since the accident. Although ^{129}I concentrations in five samples during the period of 2013-2014 have approached the pre-accident levels, ^{129}I concentrations in most samples remained higher values in winter and spring-summer. The high ^{129}I levels in winter and spring-summer are most likely attributed to local resuspension of the Fukushima-derived radionuclide bearing soil particles deposited on land surfaces, and re-emission through vegetation taking up ^{129}I from contaminated soil and water, respectively. Long-term declining rate suggests that contribution of the Fukushima-derived ^{129}I to the atmosphere would become less afterwards.

Keywords: Fukushima-derived ^{129}I, $^{129}I/^{127}I$, Long-term variation, Rainwater, Fukushima nuclear accident.

1. Introduction

After the accident of Fukushima Dai-ichi nuclear power plant (FDNPP) caused by the giant earthquake on March 11, 2011, the rainwater samples were monthly collected from Fukushima city, and analysed for ^{127}I and ^{129}I to assess the atmospheric level and behaviour of the Fukushima-derived ^{129}I (Xu et al., 2013). The ^{129}I concentration of 10^{8} atom/L in 2010

before the accident dramatically increased about four orders of magnitude to 7.6×10^{11} atom/L in March 2011 immediately after the accident with a $^{129}I/^{127}I$ ratio up to 6.9×10^{-5}. Afterwards the ^{129}I concentrations in precipitation decreased exponentially, with several fluctuations, to ~4×10^{8} atom/L in November 2012. Such a temporal variation has been explained as initial Fukushima-derived ^{129}I dispersion and re-suspension from the contaminated local environment. In comparison with the ^{129}I pre-accident level of $1.7\pm0.8\times10^{8}$ atom/L averaged from rainwater samples between November 2010 and February 2011, the value of ^{129}I concentration in November 2012 (3.6×10^{8} atom/L) remained more than two times higher. Therefore, it is necessary to understand whether the Fukushima-derived ^{129}I in atmosphere has subsequently declined to pre-accident levels. If this is a case, it is also worthwhile to investigate temporal variations since December 2012, because later elevated ^{129}I events may be an indicator of newly releases associated with the decommission of FDNPP and resuspension caused by natural processes and/or decontamination (Hirose, 2013; Tsuruta et al., 2014). This work aims to extend the investigation on levels and behaviour of Fukushima-derived ^{129}I in the atmosphere by analysis of a further 2-year time-series of precipitation samples collected from Fukushima.

2. Materials and Methods

The rainwater samples were monthly collected at campus of the Fukushima University (37°41’00”N, 140°27’16”E), located about 60 km northwest of the FDNPP. Detailed description on rainwater sample collection and analytical methods for ^{127}I and ^{129}I has been described previously (Xu et al., 2013).

A modified method was used to separation of iodine from precipitation and accelerator mass spectrometry measurement of ^{129}I. Firstly, instead of NaOH used previously, $K_2S_2O_5$ is used to treat the sample in this study in order to completely convert organic iodine in

rainwater to inorganic form (Dang et al., 2013). Secondly, instead of $^{129}I^{5+}$ ion detection at 3 MV terminal voltage in previous study, the $^{129}I^{3+}$ was chosen for detection in this study so that the ion transmission can be significantly improved for the low-level ^{129}I determination. In this operational condition, interference of $^{97}Mo^{4+}$ (disassociated from the injected MoO_2^-) to the measurement of $^{129}I^{5+}$ can be removed. Instead, there are two main interferences of $^{86}Sr^{2+}$ and $^{43}Ca^+$ (e.g. disassociated from injected $SrCO_2^-$ and $CaClO_3^-$ or $CaSO_3^-$, respectively) to $^{129}I^{3+}$, but they can be completely separated using the gas ionization detector, resulting in no need of any interference corrections.

The measured $^{129}I/^{127}I$ ratios is normalized to a standard with $^{129}I/^{127}I$ ratio of 1.098×10^{-10} prepared by dilution of the NIST 4949B standard reference material with ^{127}I carrier (Woodward) with $^{129}I/^{127}I$ ratio of 10^{-14}. The $^{129}I/^{127}I$ ratios in the prepared target of rainwater samples are in range of 10^{-12}-10^{-11}, which are more than one order of magnitude higher than those of procedure blank (10^{-13}). Repeat measurements of a secondary standard with $^{129}I/^{127}I$ ratio of 1.063×10^{-11} indicated better than 3 % precision and accuracy.

3. Results

Table 1 lists ^{127}I and ^{129}I concentrations, and $^{129}I/^{127}I$ ratios newly measured in rainwater samples collected from December 2012 to October 2014 together with previously reported ^{127}I data from March 2011 to September 2012 and ^{129}I data from November 2010 to November 2012 (Xu et al., 2013). Figure 1 shows temporal variations of ^{127}I and ^{129}I concentrations and $^{129}I/^{127}I$ ratios in the rainwater samples from the whole period from November 2010 to October 2014.

The newly measured ^{127}I concentrations between October 2012 and October 2014 varied from 0.5 μg/L in July 2014 to 10 μg/L in March 2013 with an average of 1.7±1.9 μg/L, which is consistent with the previous period between March 2011 and September 2012 ranging from

0.8 to 2.3 μg/kg with an average of 1.5±0.6 μg/L. Combination of the two datasets gives an average ^{127}I concentration of 1.6±1.5 μg/L in the period between March 2011 and October 2014. These data fall into the reported range in the literatures (0.2-12 μg/L, Aldahan et al., 2009). There is no apparent trend on temporal variation of ^{127}I concentration through the whole period.

The newly ^{129}I concentrations in rainwater samples between December 2012 and October 2014 varied from 1.2×10^{8} atoms/L in October 2014 to 6.9×10^{9} atoms/L in March 2013. The highest value is comparable with those in August-September 2011, whereas the lowest value is consistent with those observed before the accident. ^{129}I concentrations in 2012-2014 followed decline trend after the accident, but were significantly lower compared to the previous period in 2011-2012. The newly measured ^{129}I/^{127}I atomic ratios in this study vary within relatively narrow bands from 1.3×10^{-8} to 2.0×10^{-7}. The lowest ^{129}I/^{127}I ratio of 1.3×10^{-8} observed in October 2014 is consistent with those observed in pre-accident soil samples nearby (Matsunaka et al., 2015). If the average ^{127}I in the whole period 1.6±1.5 μg/L as described above can be assumed for the pre-accident rainwaters, the corresponding pre-accident ^{129}I/^{127}I atomic ratios would be $(1\text{-}4)\times10^{-8}$. Clearly, although six samples collected in the period of 2012-2014 showed ^{129}I/^{127}I ratios $(3\text{-}4)\times10^{-8}$ consistent with the pre-accident values, while ^{129}I/^{127}I ratios in most samples are 2-10 times higher than the pre-accident values.

Overall, ^{129}I and ^{129}I/^{127}I show gradually declining trends through the whole period (Figs. 1b and 1c). The whole period can be divided to four sub-periods (I: March 2011 to November 2011; II: December 2011 to November 2012; III: December 2012 to November 2013; and IV: December 2013 to October 2014), in which the decline rate of ^{129}I and ^{129}I/^{127}I in the period III and IV is obviously slower than that in the previous period I and II. A roughly seasonal variation on ^{129}I and ^{129}I/^{127}I can be observed. The high ^{129}I concentrations and ^{129}I/^{127}I ratios

123 are generally observed in winter and spring-summer, whereas the low values are found in
124 autumn.
125
126 **4. Discussion**
127 **4.1 Sources of ^{129}I in rainwaters**
128 Figure 2 shows a plot of the $^{129}I/^{127}I$ atomic ratio as a function of the ^{129}I concentration
129 measured in this work and the earlier study. At low ^{129}I concentrations (~10^8 atom/L), the
130 $^{129}I/^{127}I$ ratio approaches a value (~10^{-8}) asymptotically reflecting pre-accident atmospheric
131 levels in Fukushima. As the ^{129}I concentration increases, this ratio approaches a value that
132 reflects the ratio in Fukushima-derived component. Based on this observation, the data can be
133 fitted to a two end-member mixing equation:
134 $$[^{129}I/^{127}I]_{Meas} = [^{129}I/^{127}I]_{Bkg} + [1/^{127}I]_{Bkg} \times [^{129}I]_{Fuk} \quad (1)$$
135 where ^{129}I and $^{129}I/^{127}I$ denote ^{129}I concentrations and $^{129}I/^{127}I$ atomic ratio for sample (Meas),
136 background (Bkg) and Fukushima-derived component (Fuk). From the slope and intercept of
137 the fit, we derive an average ^{127}I concentration of 1.6 μg/L and $^{129}I/^{127}I$ ratio of 1.4×10^{-8} for
138 the pre-accident background values (Fig. 2). The ^{127}I value is in good agreement with the
139 average ^{127}I concentration of all rainwater samples through the whole period between March
140 2011 and October 2014, whereas the $^{129}I/^{127}I$ ratio is consistent with those observed in the pre-
141 accident environments in Japan which includes the atmospheric fallout in Tokyo, Tsukuba
142 and Akita (Toyama et al., 2012, 2013), surface soils observed in Fukushima (Matsunaka et
143 al., 2015) and surface soils in other non-nuclear areas (Muramatsu et al., 2008; Matsuzaki et
144 al., 2010). The level of pre-accident ^{129}I concentration and $^{129}I/^{127}I$ ratio in Japan results from
145 global fallout of weapons testing, release and dispersion from the nuclear fuel reprocessing
146 plants in Europe (in particular La Hague in France and Sellafield in UK) and in Japan (i.e.,
147 Tokai-mura), which can be considered as a local baseline of ^{129}I level (Toyama et al., 2013).

There is no doubt that source of the elevated ^{129}I exceeded the baseline of ^{129}I in Fukushima atmosphere results mainly from the ^{129}I released from the FDNPP. It is particularly true in the early period of the post-accident. With the cessation of release from the FDNPP and following rapid dispersion of Fukushima-derived radioactive plumes in atmosphere, processes causing elevated atmospheric ^{129}I in later period may include resuspension of the fine particles carrying deposited Fukushima-derived ^{129}I on land surfaces, re-emission through vegetation taking up ^{129}I from contaminated soil and water, re-emission of Fukushima-derived ^{129}I discharged and deposited in the sea, and possible releases from the damaged reactors during decommissioning.

It is difficult to assess the role of ongoing releases from the damaged reactors during the decommissioning process with the current data sets, due to the low resolution of our sampling period. Post-accident monitoring results during last four years have not shown significant new releases from the FDNPP site. Aerosol samples have been collected beside our rainwater sampling site every three days and atmospheric ^{137}Cs was monitored from May 2011 to August 2014. The measured atmospheric ^{137}Cs activities seem unlikely to suggest significant releases from the decommissioning processes of FDNPP (Watanabe, 2015). Even though there was some releases in very short period during the decommission process, they might not be captured in our monthly rainwater samples. It is nonetheless recognized that decommissioning carries potential risks of further release, and therefore it is important that sampling and monitoring should be continued.

Investigations of ^{129}I in precipitation in Europe have confirmed that ^{129}I discharged to the seas from nuclear reprocessing plants is the major source of ^{129}I in the atmosphere in Europe, especially in the North Europe (Aldahan et al., 2009). In Fukushima, in addition to gaseous release of radionuclides from the damaged reactors, indeed large amount of radioactive water has also been discharged to the sea from the FDNPP in March-April 2011, which has induced

a significant increase of the concentration of radionuclides in the coast water nearby the FDNPP. ^{129}I in seawaters in the offshore Fukushima have been reported with the highest ^{129}I concentration of 6×10^{8} atom/L and $^{129}I/^{127}I$ ratio of 2.1×10^{-9} (Hou et al., 2013). In comparison with the measured ^{129}I level in the rainwater in this study, the $^{129}I/^{127}I$ ratios in seawater are one order of magnitude lower. This seems to imply that the high dilution factors in seawater may limit the contributions of ^{129}I in the atmosphere from this pathway, although the transfer of radionuclides from sea to land via the air plays an important role on terrestrial environments along the coastal regions (McKay et al., 1990).

Figure 3 shows relationship between ^{127}I and ^{129}I concentrations in rainwater since March 2011. Overall there is no clear correlation between the two isotopes through the whole period. The similar patter is observed in the European atmosphere, which was attributed to different sources for ^{127}I and ^{129}I (Hou et al., 2009). This is particularly true for the rainwater samples soon after the accident during which ^{127}I and ^{129}I originate from different sources/reservoirs. However, if focusing on later rainwater samples from December 2012 (periods III and IV, Fig. 1c), it is found that there are somewhat positive correlations between the two isotopes as shown by best fittings (Fig. 3). Such correlations suggest a common reservoir for ^{127}I and ^{129}I with a $^{129}I/^{127}I$ atomic ratio $\sim1\times10^{-7}$ deduced from the slopes of linear correlations. It is highly considerable that this reservoir might consist of the anthropogenic Fukushima-derived ^{129}I and natural ^{127}I, which were well mixed, in environment. With the cessation of release from the FDNPP and following dispersion of Fukushima-derived ^{129}I in atmosphere with ecological half-lives of 29-44 days before October 2011 (Xu et al., 2013), the Fukushima-derived ^{129}I has deposited on land surface and sea. Miyake et al. (2015) have reported $^{129}I/^{127}I$ ratios in 50 surface soils collected in the areas within 60 km radius from FDNPP in April 2011, varying from 3×10^{-9} to 1×10^{-5} with an average of $(7\pm18)\times10^{-7}$. Muramatsu et al. (2015) investigated 82 surface soil samples within 80 km from FDNPP collected in June 2011 which have $^{129}I/^{127}I$

198 ratios from 2×10^{-8} to 4×10^{-6} with an average of $(5\pm6)\times10^{-7}$. Matsunaka et al. (2015) analysed
199 three surface soil samples nearby the FDNPP. Two samples collected 4.3 km and 8.2 km from
200 FDNPP in November 2012 showed $^{129}I/^{127}I$ ratios of 2.14×10^{-6} and 4.18×10^{-7}, respectively.
201 One sample collected 7.5 km from FDNPP in June 2013 gave $^{129}I/^{127}I$ ratio of 2.33×10^{-6}.
202 Averaging the three samples results in $^{129}I/^{127}I$ ratio of $(1.6\pm1.0)\times10^{-6}$. In addition, two
203 Japanese cedar frond samples collected in Iwaki in January 2013, 50 km southwest of the
204 FDNPP show $^{129}I/^{127}I$ ratios of 2.46×10^{-7} and 8.38×10^{-7} (Xu et al. unpublished data). These
205 datasets suggest that simple mixing of anthropogenic ^{129}I and natural ^{127}I from different
206 regions can potentially result in $^{129}I/^{127}I$ ratio of $\sim1\times10^{-7}$ in atmosphere. Therefore, it can be
207 concluded that the post-accident elevated ^{129}I in Fukushima can be mainly attributed to
208 resuspension of the fine particles bearing the deposited Fukushima-derived ^{129}I on land
209 surfaces and/or to re-emission through vegetation which uptake ^{129}I from the contaminated
210 soil and water.

211 In addition to the Fukushima-derived radionuclides, it should be pointed out that other
212 sources such as re-emission and transportation from the European reprocessing plants might
213 count for the elevated ^{129}I in this study. This may be particular true for the later periods after
214 the accident. For example, since July 2013, the ^{129}I values varied from 1.2×10^{8} atom/L to
215 4.5×10^{8} atom/L of which seven samples are below the pre-accident level. The similar results
216 are also found in five samples having $^{129}I/^{127}I$ below 5×10^{-8}, close to or below the pre-accident
217 level. Toyama et al. (2012, 2013) reported the long-term atmospheric $^{129}I/^{127}I$ ratios in Japan
218 between 1963 and 2005. In their dataset, the atmospheric $^{129}I/^{127}I$ ratios were in range of (1-
219 11)$\times10^{-8}$ in May and (0.7-3)$\times10^{-8}$ in September during the period of 1995-2005. The high
220 depositions of ^{129}I in May are attributable to the transport of the continental air mass directly
221 affected by the European sources and other possible plant located between Europe and Japan
222 (Toyama et al., 2012). Therefore, such a coincidence between Fukushima and Tokyo/Tsukuba

implies that contribution of ^{129}I from Europe cannot be ruled out for samples in spring-summer such as May-June in 2013 and 2014. To clarify these alternatives, long-term observation is required. Moreover, considering the possible spatial variations, observation near the FDNPP would be helpful to distinguish the different sources.

4.2 Factors and pathways affecting resuspension of the deposited ^{129}I

Variations of the ^{129}I concentration in atmosphere after the FDNPP accident are governed by several factors, which include the atmospheric emission history of radionuclides at the FDNPP, resuspension of radionuclide bearing soil particles, meteorological factors and migration of ^{137}Cs in land surface (Hirose, 2013). Figure 4 compares four-year variations of ^{129}I in rainwaters with ^{137}Cs in aerosols (Watanabe, 2015) both of which were collected in the same site. Overall, there is positive correction between atmospheric ^{129}I and ^{137}Cs with correlation coefficient R^2=0.9 (Fig. 4a). In fact, such a correlation can be expected because both ^{129}I and ^{137}Cs are fission products with a ^{129}I/^{137}Cs activity ratio of ~4×10^{-7} in the FDNPP (Tumey et al., 2013; Xu et al., 2015), although the two elements have distinct chemical properties in the atmosphere.

There are several ^{129}I and ^{137}Cs pulses shown by gray areas in Fig. 4a. It mainly occurred in winter season (i.e., December 2011 to February 2012, December 2012 to March 2013, December 2013 to March 2014), and spring-summer seasons (i.e., April 2012, June 2012, May 2013, May 2014). Figure 4b illustrates the monthly precipitation rate and wind speed. In comparison with meteorological factors, the radionuclides have roughly negative correlation with precipitation in 2012-2014. The relationship between radionuclides and average wind speed is not as clear as that between radionuclide and precipitation, which is probably due to frequent changes of wind directions. The similar patterns were neither seen between atmospheric radionuclides and the maximum or maximum instantaneous wind speed.

Nevertheless, it can be found that high wind speed and low precipitation roughly coincides with the high ^{129}I and ^{137}Cs concentrations. Therefore, it can be concluded that the dry and windy weather conditions favour the resuspension of the deposited Fukushima-derived radionuclides on land surfaces and accordingly elevate the ^{129}I and ^{137}Cs in atmosphere.

It has been indicated that the Fukushima-derived ^{129}I and ^{137}Cs deposited in the top surface soils are mainly bound to humus substances and clay minerals (i.e., Ohta et al., 2012; Tanaka et al., 2012; Saito et al., 2014). Furthermore, analysis of size distribution showed that both ^{129}I and ^{137}Cs dominate in fine particles (μm scale) in soils. Therefore, in dry and windy climatic conditions which are typical of winter in Fukushima (Fig. 4b), such fine soil particles can be easily suspended into the atmosphere unless large amounts of snow covers the land. This is particularly demonstrated in the rainwater sample collected in March 2013 in which both very high ^{127}I and ^{129}I are associated with the lowest precipitation and high wind speeds (Fig. 4b). In addition, dust-bearing radionuclides can be deposited on the surface of leaves of plants. When they fall to the ground and degrade in winter, such dust can also be suspended into the atmosphere. Local biomass burning, which occurs in November–December in the eastern Japan plain, is also considered to be a potential source of suspended ^{129}I and ^{137}Cs, following processed which have been recognized for Chernobyl fallout (Lujanienė et al., 2009). These observations can explain the elevated ^{129}I and ^{137}Cs concentrations observed in the winter season.

In case of the elevated radionuclides in spring-summer seasons, prevalence of biological activity might play an important role in the resuspension of ^{129}I in vegetation. MAFF (2014) has reported large amounts of radiocaesium in pollen of Japanese cedar, the most popular evergreen tree in Japan. Therefore, such radiocaesium can be released to the atmosphere with pollen in the early spring season. On the other hand, iodine isotope trace experiments have revealed that iodine volatilization from plants is a pathway for radioiodine suspension into the

atmosphere from a ^{129}I-contaminated area (Amiro and Johnston, 1989; Muramatsu and Yoshida, 1995). Although iodine emissions from living vegetation contributes ~0.1% to the ^{127}I concentration in the atmosphere above terrestrial areas (Amiro and Johnston, 1989), this pathway was thought to be a direct route for ^{129}I transport from contaminated soils to the atmosphere (Muramatsu and Yoshida, 1995). As a result, the Fukushima-derived ^{129}I can be released as gaseous form through vegetation that takes up ^{129}I from contaminated soil and water.

4.3 Long-term variation of atmospheric ^{129}I

As described above, variation of atmospheric ^{129}I and ^{129}I/^{127}I can be divided into four periods based on their decline rates (Figs. 1b and c). The average ^{129}I concentrations in periods I, II, III and IV are approximately 1×10^{11}, 2×10^{9}, 6×10^{8} and 3×10^{8} atom/L, respectively. These resulted in ^{129}I decline rates of 98%, 71% and 49% in periods II, III and IV, respectively. Similar decline rates are also observed for ^{137}Cs activities in aerosols (Watanabe, 2015).

The total amounts of suspended materials from the local surfaces might change little every year. However, radionuclides in the surface soil and forest/vegetation might gradually decrease since the accident, probably due to processes such as downward migration, resuspension and dispersion of fine particles from surface soils and forests (MAFF, 2014; Matsuda et al., 2015; Takahashi et al., 2015). Mikami et al. (2015) have constructed distribution maps of the inventories of ^{134}Cs and ^{137}Cs deposited onto ground soil using portable gamma-ray spectrometers at nearly 1000 locations within 80 km from FDNPP in March, September and December 2012. No apparent temporal change of the radionuclide inventories was observed from March to December 2012. Matsuda et al. (2015) measured ^{137}Cs activity in soil profiles in 84-85 locations with a 100 km radius from FDNPP during

three periods from December 201 to December 2012. These results suggest that radioactive cesium has moved downwards in these soils, but remained within 5 cm of the ground surface at most study sites. Takahashi et al. (2015) investigated ^{137}Cs in soil profiles under eight different land uses in July 2011, January 2012, August 2012 and December 2012. There is no clear trend observed in depth profile on any site - probably due to spatial variability not being picked up in single measurements. Honda et al. (2015) examined the variation of depth profiles from the same upland field (Kawauchi village, 20 km southwest of the FDNPP) with an interval of one year (April 2012, October 2011, December 2011 and March 2012). The downward migration rates for ^{129}I and ^{137}Cs during the interval were estimated to be 0.81±0.32 and 0.19±0.17 g/cm^2/yr, respectively. Furthermore, considering factors such as post-accident decontamination activities (i.e., removal of topmost 5 cm soils) carried out by local government, post-accident agricultural activities, and the quick downward migration which can be enhanced during the rainfall events, activity concentrations of Fukushima-derived radionuclides in the topmost soils are expected to be progressively lower. As a result, these observations suggest the gradual decreases of the ^{129}I and ^{137}Cs concentrations in the topmost soils from 2011 to 2014.

It should be pointed out that variations of ^{129}I and ^{137}Cs are not very well synchronous in 2013-2014, particularly demonstrated by the amplitude of variation. This might be attributed to the difference of baseline of ^{129}I and ^{137}Cs level in Fukushima. The atmospheric ^{137}Cs activity has been reported to be <1 μBq/m^3 in Tsukuba (Igarashi et al., 1999). This value is in good accord with the reported data in the mid-latitudes of the Northern Hemisphere. The level of ^{137}Cs in Tsukuba could be regarded as a typical value for the mid-latitudes of the Northern Hemisphere in the 1990s (Igarashi et al., 1999). This ^{137}Cs level in the air seems to be resulted from resuspension of the global fallout from atmospheric nuclear bomb tests, etc. and can be regarded as the background of atmospheric ^{137}Cs in Japan. In comparison with this

background level, the Fukushima atmospheric ^{137}Cs concentrations in 2013-2014 (10-100 μBq/m^3) are still 10-100 times higher. In contrast, large amounts of ^{129}I discharged from the nuclear reprocessing facilities in Europe have resulted in largely elevated ^{129}I in global atmosphere (Aldahan et al., 2009), although the present ^{137}Cs concentration in the European air is <1 μBq/m^3 (i.e., Nielsen et al., 2015). This added ^{129}I flux has caused enhancement of baseline of global ^{129}I level (i.e., ^{129}I/^{127}I>10-7 in Denmark, Hou et al., 2009). As a result, the Fukushima ^{129}I concentrations in 2013-2014 are only up to 4 times higher than the pre-accident level, except for one sample in March 2013, which shows typical resuspension of local Fukushima-derived ^{129}I, and natural ^{127}I in land surface. Thus, as a result of higher background level of ^{129}I in the atmosphere, with the gradually decreasing of Fukushima-derived ^{129}I in the suspended matter, effect of Fukushima-derived ^{129}I might become less contribution to the present ^{129}I level in respective with the ^{137}Cs afterwards.

5. Conclusions

Variations of iodine isotopes in four-year time series rainwater collected in Fukushima, Japan over 2010-2014 reveal that Fukushima-derived ^{129}I has gradually declined from 7.6×10^{11} atom/L in March 2011 immediately after the accident to $(1\text{-}4)\times10^{8}$ atom/L in 2014, having intermittently approached the pre-accident level $(1\text{-}3)\times10^{8}$ atom/L. The atmospheric ^{129}I and ^{137}Cs showed roughly a season variation, high values in winter and low in autumn. Enhancement of ^{129}I during the studied period can be mainly attributed to resuspension of the Fukushima-derived radionuclide bearing particles deposited on land surfaces under the winter dry and windy climatic conditions, and re-emission through vegetation taking up ^{129}I from contaminated soil and water in spring-summer. However, possibility of ^{129}I transported from the European reprocessing plants cannot be ruled out. Long-term declining rate suggests that contribution of the Fukushima-derived ^{129}I to the atmosphere would become less afterwards.

Acknowledgements

We thank A. Dougass and P. Gallacher of SUERC for assistance in AMS measurement. The Great Britain Sasakawa Foundation is appreciated for partial support in sample collection.

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Figure captions

Figure 1. Temporal variations of ^{127}I (a), ^{129}I (b) and $^{129}I/^{127}I$ (c) in rainwater samples from Fukushima. The open circles denote samples with the averaged ^{127}I (1.6 μg/L) of the whole period and the corresponding $^{129}I/^{127}I$ ratio

Figure 2. Correlation between ^{129}I and $^{129}I/^{127}I$ in rainwater samples from Fukushima

Figure 3. Correlation between ^{127}I and ^{129}I in rainwater samples from Fukushima

Figure 4. (a) Comparison of temporal variation of ^{129}I in rainwaters and ^{137}Cs in aerosols from Fukushima. The monthly ^{137}Cs data were averaged from the measured values of every three days (Watanabe, 2015). (b) variation of average monthly precipitation and wind speed in Fukushima (data sources: http://www.jma.go.jp/jma/index.html)

Figure 1

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Figure 1

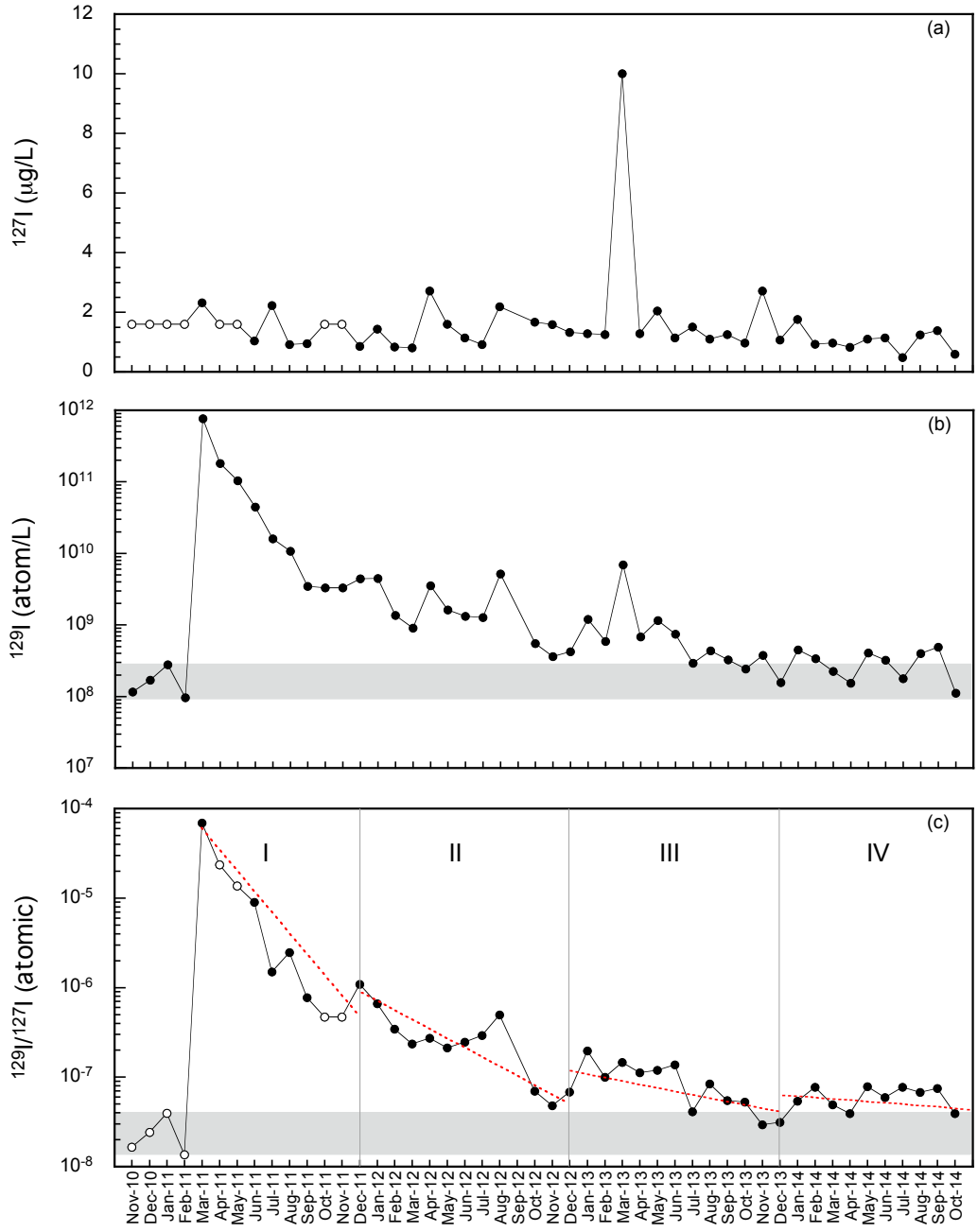

Figure 2
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Figure 2

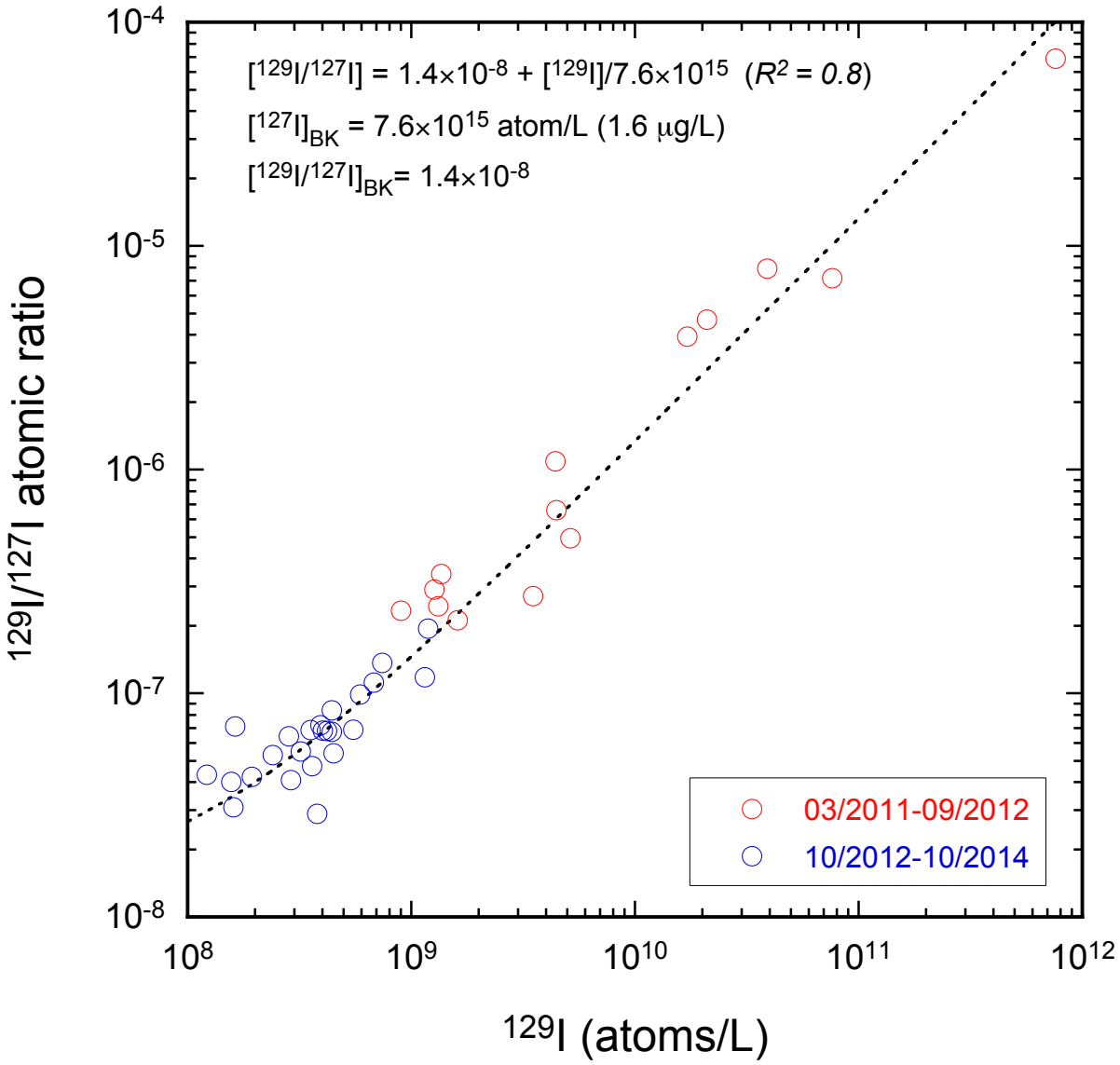

Figure 3
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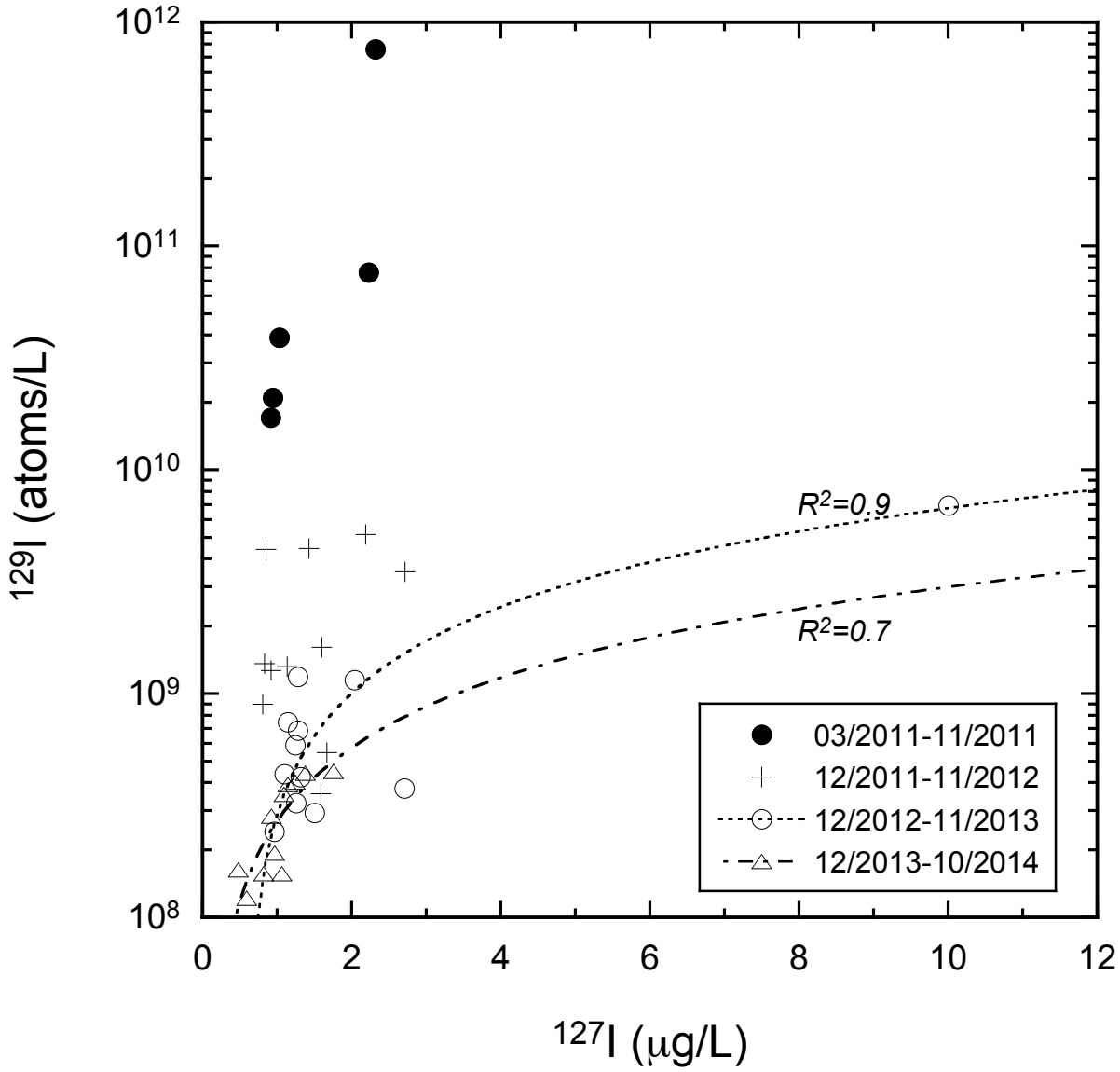

Figure 4

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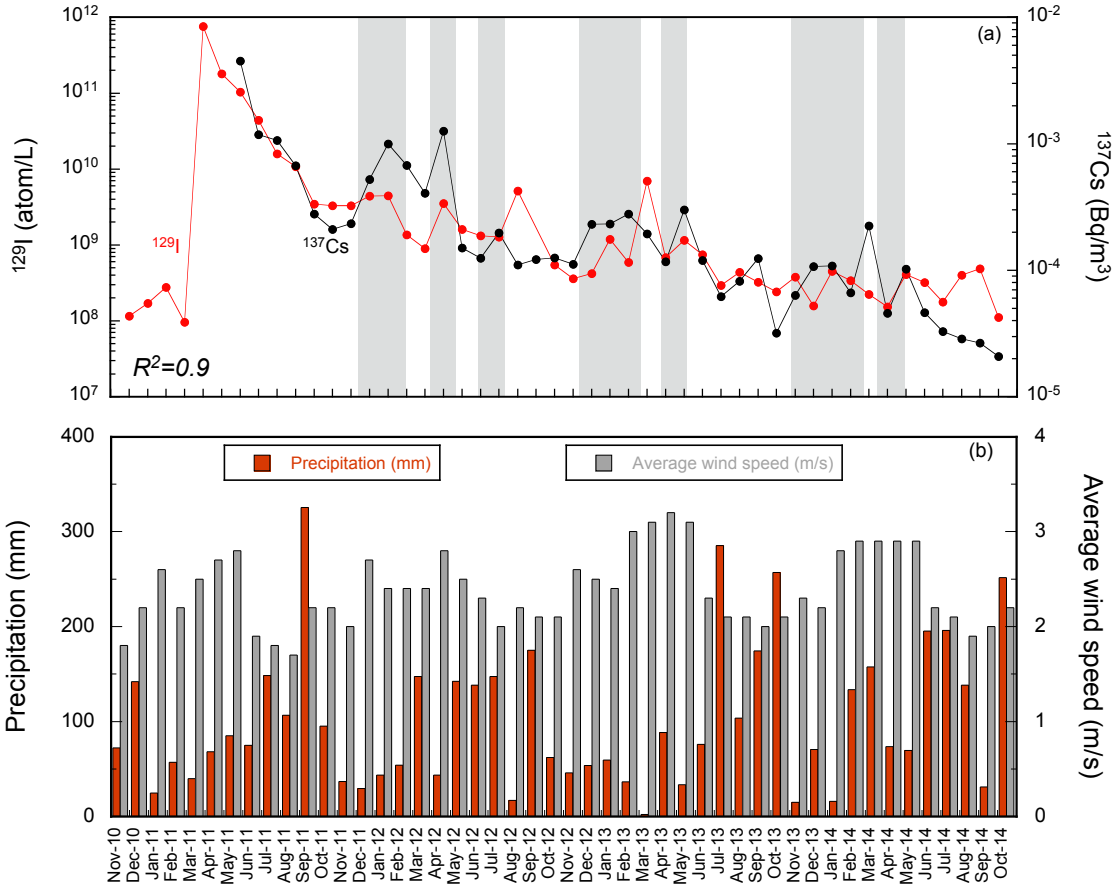

Table 1

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Table 1. ^{127}I and ^{129}I concentrations in rainwaters collected from Fukushima*

Sampling date	^{127}I (μg/L)	^{129}I ($\times 10^{8}$ atom/L)	$^{129}I/^{127}I$ ($\times 10^{-8}$, atomic)
Nov 2010		1.16±0.10	
Dec 2010		1.70±0.10	
Jan 2011		2.77±0.10	
Feb 2011		0.96±0.10	
Mar 2011	2.321±0.018	7584.08±199.48	6890.05±189.04
1-20 Apr 2011		1789.94±20.75	
21 Apr - 9 May 2011		1037.14±28.00	
3 Jun 2011	1.036±0.012	389.92±9.30	793.4±21.08
15 Jun 2011	2.230±0.014	761.82±17.49	720.31±17.16
30 Jun 2011	0.917±0.002	171.28±1.12	393.81±2.72
15 Jul 2011	0.943±0.004	209.77±3.40	468.89±7.87
16 Jul - 5 Aug 2011		108.05±1.16	
Aug 2011		107.41±1.18	
Sep 2011		34.49±0.60	
Oct 2011		32.94±0.70	
Dec 2011	0.854±0.014	44.25±0.49	109.19±2.18
Jan 2012	1.430±0.018	44.61±0.49	65.77±1.10
Feb 2012	0.836±0.010	13.56±0.22	34.20±0.70
Mar 2012	0.808±0.014	8.98±0.20	23.44±0.67
Apr 2012	2.717±0.016	35.10±0.38	27.24±0.33
May 2012	1.600±0.016	16.09±0.24	21.21±0.39
Jun 2012	1.137±0.012	13.21±0.23	24.49±0.50
Jul 2012	0.921±0.010	12.74±0.21	29.17±0.58
Aug 2012	2.188±0.016	51.29±0.60	49.44±0.69
Oct 2012	1.669±0.012	5.45±0.10	6.88±0.14
Nov 2012	1.590±0.011	3.57±0.10	4.73±0.14
Dec 2012	1.318±0.007	4.23±0.27	6.76±0.44
Jan 2013	1.281±0.002	11.87±0.22	19.54±0.37
Feb 2013	1.247±0.002	5.88±0.12	9.94±0.20
Mar 2013	10.005±0.054	69.35±1.59	14.62±0.34
Apr 2013	1.281±0.012	6.83±0.12	11.24±0.23
May 2013	2.041±0.014	11.47±0.19	11.85±0.21
Jun 2013	1.144±0.010	7.45±0.13	13.72±0.27
Jul 2013	1.506±0.027	2.93±0.07	4.10±0.13
Aug 2013	1.100±0.003	4.37±0.09	8.38±0.18
Sep 2013	1.256±0.007	3.25±0.08	5.46±0.13
Oct 2013	0.966±0.004	2.42±0.07	5.29±0.14
Nov 2013	2.709±0.015	3.77±0.13	2.93±0.10
Dec 2013	1.065±0.005	1.57±0.06	3.10±0.12
Jan 2014	1.759±0.005	4.49±0.11	5.39±0.13
Feb 2014	0.927±0.005	2.83±0.20	6.44±0.47
Mar 2014	0.968±0.008	1.94±0.12	4.23±0.26
Apr 2014	0.824±0.005	1.57±0.10	4.02±0.27
May 2014	1.094±0.008	3.55±0.15	6.86±0.29
Jun 2014	1.146±0.005	3.92±0.39	7.21±0.73
Jul 2014	0.483±0.006	1.63±0.11	7.11±0.51
Aug 2014	1.244±0.007	4.02±0.16	6.82±0.28
Sep 2014	1.384±0.006	4.42±0.19	6.74±0.30
Oct 2014	0.596±0.010	1.22±0.12	4.33±0.43

*^{127}I and $^{129}I/^{127}I$ data prior to Oct 2012 and ^{129}I data prior to Dec 2012 from Xu et al. (2013)